

# Excess Molar Volumes and Surface Tensions of 1,2,4-Trimethylbenzene and 1,3,5-Trimethylbenzene with 1-Butanol, 2-Methyl-1-propanol, 2-Butanol, and 2-Methyl-2-propanol at 298.15 K

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Densities for binary mixtures of (1,2,4-trimethylbenzene + 1-butanol, 2-methyl-1-propanol, 2-butanol, and 2-methyl-2-propanol and 1,3,5-trimethylbenzene + 1-butanol, 2-methyl-1-propanol, 2-butanol, and 2-methyl-2-propanol) have been determined over the entire concentration range at 298.15 K, and excess molar volumes have been derived. Surface tensions of these binary mixtures have been measured at 298.15 K by the pendant drop method, and the values of the surface tension deviation for these mixtures were also calculated.

## Introduction

The surface tension and density of liquids and liquid mixtures are important physical properties because they play an important role in the mass and heat transfer at an interface such as in liquid–liquid extraction, gas absorption, distillation, and condensation. One of our research directions is xylene and trimethylbenzene separation. In previous papers, the excess molar volumes and surface tensions at 298.15 K for xylene + alkanol (2-propanol and 2-methyl-2-propanol), xylene + alkone (acetone and 2-butanone), and xylene + ether (isopropyl ether and methyl *tert*-butyl ether) systems were reported.<sup>1–3</sup> In this paper, surface tensions and densities for 1,2,4-trimethylbenzene + 1-butanol, 2-methyl-1-propanol, 2-butanol, or 2-methyl-2-propanol and 1,3,5-trimethylbenzene + 1-butanol, 2-methyl-1-propanol, 2-butanol, or 2-methyl-2-propanol at 298.15 K are reported.

## Experimental Section

1,2,4-Trimethylbenzene (ACROS Organics), 1,3,5-trimethylbenzene (ACROS Organics), 1-butanol (GuangZhou Chem., China), 2-methyl-1-propanol (ShangHai Chem., China), 2-butanol (ACROS Organics), and 2-methyl-2-propanol (TianJin Chem., China) were of high grade. The mass fraction purities of the substances were 1,2,4-trimethylbenzene (99.20%), 1,3,5-trimethylbenzene (99.30%), 1-butanol (99.86%), 2-methyl-1-propanol (99.88%), 2-butanol (99.90%), and 2-methyl-2-propanol (99.80%), as determined by a PE auto system XL gas chromatograph. All of the chemicals were prepared by a molecular sieve treatment, and all of the mixtures were measured by mass using an Ohaus E12140 balance with an accuracy of  $\pm 0.1$  mg.

Densities of the pure liquids and their mixture were measured with an Anton Paar DMA 4500 vibrating tube densimeter thermostated at  $(298.15 \pm 0.01)$  K. The densimeter error was  $\pm 5 \times 10^{-5}$  g·cm<sup>-3</sup>. The surface tensions of the pure liquids and their mixtures were determined by the pendant drop method using a Dataphysics OCA20

**Table 1. Physical Properties of the Pure Components at 298.15 K**

substance	$\rho/\text{g}\cdot\text{cm}^{-3}$		$\sigma/\text{mN}\cdot\text{m}^{-1}$	
	exptl	lit	exptl	lit
1,2,4-trimethylbenzene	0.87164	0.87174 <sup>a</sup>	29.25	29.19 <sup>d</sup>
1,3,5-trimethylbenzene	0.86103	0.86109 <sup>b</sup>	28.09	27.54 <sup>d</sup>
1-butanol	0.80580	0.80575 <sup>c</sup>	24.18	24.93 <sup>d</sup>
2-methyl-1-propanol	0.79783	0.79790 <sup>c</sup>	22.30	22.54 <sup>d</sup>
2-butanol	0.80234	0.80241 <sup>f</sup>	23.46	22.62 <sup>30,d</sup>
				22.14 <sup>30</sup>
2-methyl-2-propanol	0.78085	0.78120 <sup>g</sup>	20.13	20.10 <sup>g</sup>

<sup>a</sup> Reference 7. <sup>b</sup> Reference 8. <sup>c</sup> Reference 9. <sup>d</sup> Reference 10. <sup>e</sup> Reference 11. <sup>f</sup> Reference 12. <sup>g</sup> Reference 13.

contact angle and surface tension measuring device. This instrument provides a computer-controlled display (CCD) video camera to take pictures and an electronic syringe unit to inject samples, so the surface tension of the sample can be determined very quickly. The surface tension is given by<sup>4</sup>

$$\sigma = \frac{g \Delta \rho d_e^2}{H} \quad (1)$$

where  $g$  is the gravitational acceleration,  $\Delta \rho$  is the density difference between the droplet and the surrounding,  $d_e$  is the largest diameter of the drop, and  $H$  is a correction factor that depends on the sharpness of the drop. The error of the instrument is  $\pm 0.05$  mN·m<sup>-1</sup> ( $\pm 0.1$  K). The densities and surface tensions of the pure compounds are given in Table 1 and compared with the literature values.

## Result and Discussion

Excess molar volumes were determined from the density data<sup>5</sup>

$$V^E = \frac{x_1 M_1 + x_2 M_2}{\rho} - \frac{M_1}{x_1 \rho_1} - \frac{M_2}{x_2 \rho_2} \quad (2)$$

where  $M_i$  represents the molar mass of component  $i$ ,  $\rho$  and  $\rho_i$  are the densities of the mixture and component  $i$ ,

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**Table 2.** Experimental Excess Molar Volumes  $V^E$  at 298.15 K

$x_1$	$V^E$ $\text{cm}^3 \cdot \text{mol}^{-1}$	$x_1$	$V^E$ $\text{cm}^3 \cdot \text{mol}^{-1}$	$x_1$	$V^E$ $\text{cm}^3 \cdot \text{mol}^{-1}$	$x_1$	$V^E$ $\text{cm}^3 \cdot \text{mol}^{-1}$
$x\text{CH}_3\text{CH}_2\text{CH}_2\text{CH}_2\text{OH} + (1-x)1,2,4-\text{C}_6\text{H}_3(\text{CH}_3)_3$							
0.0499	0.048	0.2992	0.164	0.5498	0.120	0.8001	0.035
0.1013	0.101	0.3503	0.162	0.5985	0.105	0.8501	0.021
0.1492	0.130	0.4009	0.154	0.6501	0.087	0.9002	0.009
0.2002	0.149	0.4491	0.145	0.6987	0.070	0.9503	0.003
0.2499	0.160	0.5012	0.132	0.7499	0.051		
$x\text{CH}_3\text{CH}_2\text{CH}_2\text{CH}_2\text{OH} + (1-x)1,3,5-\text{C}_6\text{H}_3(\text{CH}_3)_3$							
0.0493	0.077	0.3002	0.211	0.5484	0.186	0.7999	0.095
0.1006	0.132	0.3487	0.213	0.6005	0.172	0.8500	0.074
0.1512	0.169	0.4003	0.211	0.6500	0.158	0.9010	0.047
0.2011	0.190	0.4535	0.206	0.6993	0.137	0.9506	0.019
0.2514	0.203	0.5001	0.198	0.7497	0.118		
$x\text{CH}_3\text{CH}(\text{CH}_3)\text{CH}_2\text{OH} + (1-x)1,2,4-\text{C}_6\text{H}_3(\text{CH}_3)_3$							
0.0509	0.093	0.3002	0.272	0.5488	0.278	0.8002	0.146
0.1004	0.162	0.3487	0.286	0.6002	0.258	0.8506	0.109
0.1530	0.205	0.3996	0.295	0.6508	0.237	0.8984	0.071
0.2018	0.235	0.4494	0.297	0.6989	0.210	0.9496	0.033
0.2490	0.254	0.4995	0.289	0.7484	0.180		
$x\text{CH}_3\text{CH}(\text{CH}_3)\text{CH}_2\text{OH} + (1-x)1,3,5-\text{C}_6\text{H}_3(\text{CH}_3)_3$							
0.0512	0.114	0.3006	0.326	0.5480	0.344	0.7998	0.217
0.1028	0.183	0.3529	0.342	0.6002	0.327	0.8504	0.172
0.1517	0.233	0.4019	0.356	0.6495	0.312	0.9006	0.122
0.1994	0.273	0.4494	0.359	0.6990	0.286	0.9485	0.064
0.2514	0.301	0.4995	0.358	0.7501	0.250		
$x\text{CH}_3\text{CH}_2\text{CH}(\text{CH}_3)\text{OH} + (1-x)1,2,4-\text{C}_6\text{H}_3(\text{CH}_3)_3$							
0.0497	0.127	0.2985	0.389	0.5486	0.385	0.7999	0.221
0.1008	0.223	0.3492	0.408	0.6008	0.362	0.8496	0.171
0.1486	0.282	0.4008	0.416	0.6505	0.332	0.9002	0.116
0.2010	0.332	0.4506	0.410	0.6994	0.295	0.9489	0.063
0.2507	0.368	0.4997	0.402	0.7503	0.264		
$x\text{CH}_3\text{CH}_2\text{CH}(\text{CH}_3)\text{OH} + (1-x)1,3,5-\text{C}_6\text{H}_3(\text{CH}_3)_3$							
0.0508	0.144	0.2986	0.460	0.5484	0.486	0.8009	0.294
0.1010	0.250	0.3488	0.484	0.6004	0.462	0.8504	0.234
0.1511	0.326	0.4007	0.500	0.6508	0.433	0.9002	0.160
0.2007	0.387	0.4495	0.504	0.6991	0.398	0.9505	0.074
0.2500	0.424	0.5017	0.501	0.7496	0.353		
$x(\text{CH}_3)_3\text{COH} + (1-x)1,2,4-\text{C}_6\text{H}_3(\text{CH}_3)_3$							
0.0498	0.165	0.3001	0.497	0.5494	0.613	0.7993	0.374
0.1000	0.269	0.3510	0.536	0.6006	0.595	0.8500	0.296
0.1496	0.348	0.4002	0.562	0.6502	0.566	0.9003	0.197
0.2007	0.409	0.4496	0.590	0.6991	0.509	0.9499	0.101
0.2495	0.453	0.5005	0.607	0.7491	0.451		
$x(\text{CH}_3)_3\text{COH} + (1-x)1,3,5-\text{C}_6\text{H}_3(\text{CH}_3)_3$							
0.0502	0.168	0.2995	0.594	0.5483	0.751	0.7995	0.538
0.1015	0.298	0.3484	0.645	0.6005	0.747	0.8501	0.432
0.1482	0.386	0.3995	0.686	0.6502	0.730	0.9002	0.315
0.2007	0.472	0.4490	0.717	0.6987	0.692	0.9509	0.155
0.2509	0.537	0.4992	0.746	0.7500	0.627		

**Table 3.** Least-Squares Parameters and Standard Deviations

	$A_0$	$A_1$	$A_2$	$A_3$	$A_4$	$s^a$
1-butanol + 1,2,4-trimethylbenzene	0.5313	0.5486	0.1816	0.1012	-0.1439	0.0024
1-butanol + 1,3,5-trimethylbenzene	0.7893	0.3787	0.2516	0.3253	0.0970	0.0015
2-methyl-1-propanol + 1,2,4-trimethylbenzene	1.1536	0.2835	-0.1140	0.4977	0.4710	0.0038
2-methyl-1-propanol + 1,3,5-trimethylbenzene	1.4216	0.1933	0.0998	0.3275	0.4877	0.0033
2-butanol + 1,2,4-trimethylbenzene	1.6066	0.5012	0.1815	0.2967	0.3743	0.0029
2-butanol + 1,3,5-trimethylbenzene	1.9944	0.3027	0.2928	0.4706	0.1770	0.0033
2-methyl-2-propanol + 1,2,4-trimethylbenzene	2.4295	-0.3429	-0.2572	1.3514	0.8539	0.0033
2-methyl-2-propanol + 1,3,5-trimethylbenzene	2.9728	-0.7036	-0.4664	0.9346	0.1696	0.0034

<sup>a</sup> Given in units of  $\text{cm}^3 \cdot \text{mol}^{-1}$ .

and  $x_i$  is the molar fraction of component  $i$ . Experimental excess molar volumes  $V^E$  for eight binary mixtures (1,2,4-trimethylbenzene + 1-butanol, 2-methyl-1-propanol, 2-butanol, or 2-methyl-2-propanol and 1,3,5-trimethylbenzene + 1-butanol, 2-methyl-1-propanol, 2-butanol, or 2-methyl-2-propanol) at 298.15 K are listed in Table 2 and graphically presented in Figure 1. The experimental results were fit by the method of least

squares with all points weighted equally to the smoothing equation<sup>6</sup>

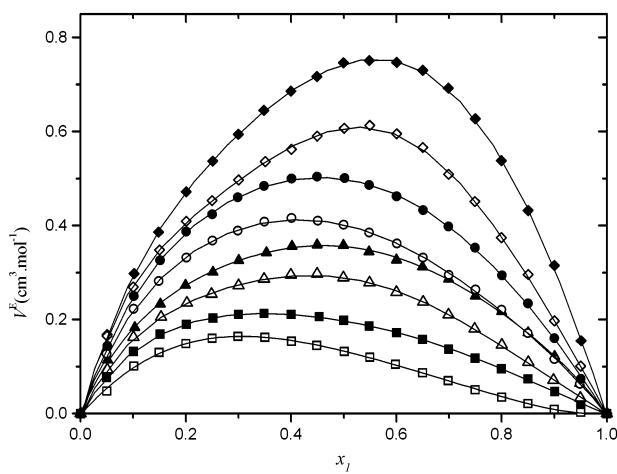
$$V^E = x(1-x) \sum_{i=0}^k A_i (1-2x)^i \quad (3)$$

The parameters  $A_0, A_1, A_2, A_3$ , and  $A_4$  and the standard deviations are given in Table 3. Table 4 lists the surface

**Table 4.** Surface Tensions  $\sigma$  at 298.15 K<sup>a</sup>

$x_1$	$\sigma$	$\delta\sigma$	$x_1$	$\sigma$	$\delta\sigma$	$x_1$	$\sigma$	$\delta\sigma$	$x_1$	$\sigma$	$\delta\sigma$		
0.1013	$x\text{CH}_3\text{CH}_2\text{CH}_2\text{CH}_2\text{OH} + (1-x)\text{1,2,4-C}_6\text{H}_3(\text{CH}_3)_3$	28.81	0.07	0.5985	26.65	0.43	0.1006	$x\text{CH}_3\text{CH}_2\text{CH}_2\text{CH}_2\text{OH} + (1-x)\text{1,3,5-C}_6\text{H}_3(\text{CH}_3)_3$	27.92	0.22	0.5001	25.99	0.25
0.2002		28.42	0.19	0.6987	26.08	0.37	0.2011		27.65	0.35	0.6005	25.53	0.17
0.2992		28.04	0.31	0.8001	25.49	0.30	0.3002		27.33	0.41	0.6993	25.05	0.09
0.4009		27.61	0.39	0.9002	24.86	0.17	0.4003		26.91	0.39	0.7999	24.58	0.01
0.5012		27.15	0.44				0.5001		26.46	0.33			
0.1004	$x\text{CH}_3\text{CH}(\text{CH}_3)\text{CH}_2\text{OH} + (1-x)\text{1,2,4-C}_6\text{H}_3(\text{CH}_3)_3$	28.42	-0.13	0.6002	24.72	-0.36	0.1028	$x\text{CH}_3\text{CH}(\text{CH}_3)\text{CH}_2\text{OH} + (1-x)\text{1,3,5-C}_6\text{H}_3(\text{CH}_3)_3$	27.65	0.16	0.6002	24.88	0.27
0.2018		27.54	-0.31	0.6989	24.16	-0.23	0.1994		27.21	0.27	0.6990	24.25	0.21
0.3002		26.70	-0.46	0.8002	23.60	-0.09	0.3006		26.67	0.32	0.7998	23.61	0.15
0.3996		25.95	-0.52	0.8984	22.98	-0.03	0.4019		26.09	0.33	0.9006	22.97	0.09
0.4995		25.29	-0.49				0.4995		25.51	0.31			
0.1008	$x\text{CH}_3\text{CH}_2\text{CH}(\text{CH}_3)\text{OH} + (1-x)\text{1,2,4-C}_6\text{H}_3(\text{CH}_3)_3$	28.58	-0.09	0.6008	25.59	-0.18	0.1010	$x\text{CH}_3\text{CH}_2\text{CH}(\text{CH}_3)\text{OH} + (1-x)\text{1,3,5-C}_6\text{H}_3(\text{CH}_3)_3$	27.77	0.15	0.6004	25.49	0.18
0.2010		27.94	-0.15	0.6994	25.07	-0.13	0.2007		27.36	0.20	0.6991	24.98	0.13
0.2985		27.32	-0.20	0.7999	24.56	-0.06	0.2986		26.94	0.23	0.8009	24.45	0.07
0.4008		26.71	-0.22	0.9002	24.02	-0.02	0.4007		26.47	0.24	0.9002	23.94	0.02
0.4997		26.15	-0.21				0.5017		25.99	0.22			
0.1000	$x(\text{CH}_3)_3\text{COH} + (1-x)\text{1,2,4-C}_6\text{H}_3(\text{CH}_3)_3$	28.12	-0.22	0.6006	23.23	-0.54	0.1015	$x(\text{CH}_3)_3\text{COH} + (1-x)\text{1,3,5-C}_6\text{H}_3(\text{CH}_3)_3$	27.18	-0.10	0.6005	23.02	-0.29
0.2007		27.00	-0.42	0.6991	22.46	-0.41	0.2007		26.24	-0.25	0.6987	22.32	-0.21
0.3001		25.92	-0.59	0.7993	21.70	-0.26	0.2995		25.35	-0.36	0.7995	21.61	-0.12
0.4002		24.93	-0.67	0.9003	20.96	-0.08	0.3995		24.51	-0.40	0.9002	20.86	-0.06
0.5005		24.06	-0.63				0.4992		23.75	-0.37			

<sup>a</sup>  $\sigma$  and  $\delta\sigma$  are given in units of  $\text{mN}\cdot\text{m}^{-1}$ .

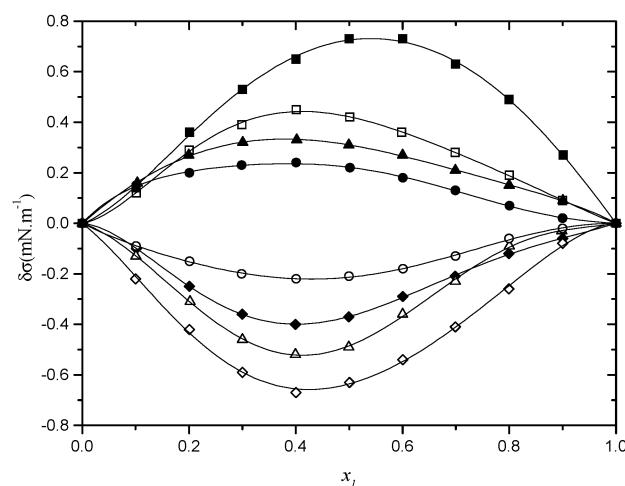


**Figure 1.** Excess molar volumes  $V^E$  for  $(x)n\text{-butanol}$ :  $\square$ ,  $+(1-x)\text{1,2,4-trimethylbenzene}$ ;  $\blacksquare$ ,  $+(1-x)\text{1,3,5-trimethylbenzene}$ .  $(x)\text{iso}$ -butanol:  $\triangle$ ,  $+(1-x)\text{1,2,4-trimethylbenzene}$ ;  $\blacktriangle$ ,  $+(1-x)\text{1,3,5-trimethylbenzene}$ .  $(x)\text{sec}$ -butanol:  $\circ$ ,  $+(1-x)\text{1,2,4-trimethylbenzene}$ ;  $\bullet$ ,  $+(1-x)\text{1,3,5-trimethylbenzene}$ .  $(x)\text{tert}$ -butyl alcohol:  $\diamond$ ,  $+(1-x)\text{1,2,4-trimethylbenzene}$ ;  $\blacklozenge$ ,  $+(1-x)\text{1,3,5-trimethylbenzene}$  at 298.15 K.

tensions and surface tension deviations for eight binary mixtures ( $1,2,4$ -trimethylbenzene + 1-butanol, 2-methyl-1-propanol, 2-butanol, or 2-methyl-2-propanol and  $1,3,5$ -trimethylbenzene + 1-butanol, 2-methyl-1-propanol, 2-butanol, or 2-methyl-2-propanol) at 298.15 K. The surface tension deviations  $\delta\sigma$  are defined by<sup>5</sup>

$$\delta\sigma = \sigma - x_1\sigma_1 - x_2\sigma_2 \quad (4)$$

The result of  $V^E$  values at 298.15 K from Figure 1 shows that they are all positive for these eight binary mixtures. The maximum values of  $V^E$  for them follow the order 1-butanol +  $1,2,4$ -trimethylbenzene < 1-butanol +  $1,3,5$ -trimethylbenzene < 2-methyl-1-propanol +  $1,2,4$ -trimethylbenzene < 2-methyl-1-propanol +  $1,3,5$ -trimethylbenzene < 2-butanol +  $1,2,4$ -trimethylbenzene < 2-butanol +  $1,3,5$ -trimethylbenzene < 2-methyl-2-propanol +  $1,2,4$ -trimethylbenzene < 2-methyl-2-propanol +  $1,3,5$ -trimethylbenzene.



**Figure 2.** Surface tension deviations  $\delta\sigma$  for  $(x)n\text{-butanol}$ :  $\square$ ,  $+(1-x)\text{1,2,4-trimethylbenzene}$ ;  $\blacksquare$ ,  $+(1-x)\text{1,3,5-trimethylbenzene}$ .  $(x)i\text{-butanol}$ :  $\triangle$ ,  $+(1-x)\text{1,2,4-trimethylbenzene}$ ;  $\blacktriangle$ ,  $+(1-x)\text{1,3,5-trimethylbenzene}$ .  $(x)\text{sec}$ -butanol:  $\circ$ ,  $+(1-x)\text{1,2,4-trimethylbenzene}$ ;  $\bullet$ ,  $+(1-x)\text{1,3,5-trimethylbenzene}$ .  $(x)t\text{-butyl alcohol}$ :  $\diamond$ ,  $+(1-x)\text{1,2,4-trimethylbenzene}$ ;  $\blacklozenge$ ,  $+(1-x)\text{1,3,5-trimethylbenzene}$  at 298.15 K.

ylbenzene < 2-methyl-2-propanol +  $1,3,5$ -trimethylbenzene. It is obvious that the packing effect plays a more important role than the association between aromatic molecules and the hydroxide radical of the four kinds of alcohol molecules, which results in positive  $V^E$  values. Because 2-methyl-2-propanol has a stronger packing effect than the other molecules, the  $V^E$  values of 2-methyl-2-propanol + trimethylbenzene are more positive than those of 1-butanol, 2-methyl-1-propanol, or 2-butanol + trimethylbenzene.

Figure 2 shows that the surface tension deviations  $\delta\sigma$  at 298.15 K are positive for mixtures of 1-butanol +  $1,3,5$ -trimethylbenzene, 2-methyl-1-propanol +  $1,3,5$ -trimethylbenzene, 2-butanol +  $1,3,5$ -trimethylbenzene, and 1-butanol +  $1,2,4$ -trimethylbenzene. The maximum values (at about  $x = 0.4$  or 0.5) of  $\delta\sigma$  for them follow the order

2-butanol + 1,3,5-trimethylbenzene < 2-methyl-1-propanol + 1,3,5-trimethylbenzene < 1-butanol + 1,2,4-trimethylbenzene < 1-butanol + 1,3,5-trimethylbenzene. It also shows that the surface tension deviations  $\delta\sigma$  at 298.15 K are negative for mixtures of 2-methyl-2-propanol + 1,2,4-trimethylbenzene, 2-methyl-1-propanol + 1,2,4-trimethylbenzene, 2-methyl-2-propanol + 1,3,5-trimethylbenzene, and 2-butanol + 1,2,4-trimethylbenzene. The minimum values (at about  $x = 0.4$ ) of  $\delta\sigma$  follow the order 2-methyl-2-propanol + 1,2,4-trimethylbenzene < 2-methyl-1-propanol + 1,2,4-trimethylbenzene < 2-butanol + 1,2,4-trimethylbenzene.

The surface tension deviation  $\delta\sigma$  can be considered to result from the following: One is association between an aromatic molecule and the hydroxide radical of butanol, one is a packing effect, and another is a dipolar–dipolar interaction. For the binary systems with positive deviation, the  $\sigma-\pi$  bond formed between an aromatic molecule and the hydroxide radical of butanol is more dominant than the packing effect. Conversely, for the binary systems with negative deviation, because 2-methyl-2-propanol is a large molecule with steric hindrance, the packing effect is more dominant than the  $\sigma-\pi$  bond formed by an aromatic molecule and the hydroxide radical of butanol, which results in a decrease in surface tension.

#### Supporting Information Available:

Density data of 1,2,4-trimethylbenzene and 1,3,5-trimethylbenzene with *n*-butanol, *iso*-butanol, *sec*-butanol, and *tert*-butanol at 298.15 K. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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